On the Viscosity of Natural Gases from Qatari North Field Reservoir

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New viscosity measurements of three natural gas-like mixtures with compositions resembling those of the Qatari North Field reservoir are reported in this work. The viscosity measurements were performed with an electromagnetic piston viscometer for temperatures from (250 to 450) K and pressure between (10 and 65) MPa. The apparatus was calibrated by use of nitrogen and propane as viscosity reference data. Uncertainties of reported data are ± 2.5 % for pressure less than 30 MPa and ± 4 % for higher pressures. The predictive ability of several models was analyzed in relation to the experimental reported viscosity data, leading to large deviations from the reported measurements. The effects of mixtures' composition on viscosity were also studied.

Introduction

The total world production of natural gas is expected to increase from the 3.107 · 10^{12} m³ produced in 2008 to 4.370 · 10^{12} m³ in 2035, according to the most reasonable scenarios, and thus, a 1.4 % annual average percent change would be required.[1] The economic importance of natural gas may be inferred by considering that 39 % and 33 % of the world production is used for industrial purposes and for electricity generation, respectively.[1] Moreover, when carbon dioxide emissions from fossil fuel combustion are considered, it is clear that natural gas should be considered as a cleaner fuel in comparison with oil and coal.[2]

The probed world total natural gas reserves are estimated to be 185.8 · 10^{12} m³ in 2010, which is a remarkable increment of 9.956 · 10^{12} m³ when compared with 2009 data.[3] The analysis of probed reserves by country shows that Qatar, with 25.28 · 10^{12} m³ probed, has the world's third largest reservoirs, and thus, Qatar holds almost 14 % of total world natural gas.[3] In 2008, Qatar produced 75.9 · 10^{9} m³ of natural gas,[1] which is more than 5 times the amount produced in 1995; nevertheless, this quantity will increase remarkably in the next years.[1] The majority of Qatar's natural gas is located in the massive offshore North Field, the world's largest nonassociated natural gas field. The plans to expand natural gas production from North Field, to meet the expected increase in regional demand and to supply markets outside the region, would allow North Field to produce 205.32 · 10^{9} m³·a^{-1} in 2012.[4] Increasing Qatar's natural gas production will lead to large-scale projects such as new liquefied natural gas (LNG) infrastructure (Qatar is the first world LNG exporter), natural gas exports through the Dolphin pipeline, several large-scale gas-to-liquids (GTL) projects, and the promotion of downstream industries that utilize natural gas as feedstock.[5]

The increasing worldwide demand for natural gas is leading to the need for developing reliable and accurate reservoir characterization and simulation. The upstream gas industry is also being faced with increasing needs for precision in the monitoring of gas supplies. Therefore, for optimal production, processing, transportation, and usage of natural gas, accurate and reliable knowledge of the natural gas viscosity, along with other thermophysical properties, is a prerequisite. The effect of gas viscosity estimation errors on the gas recovery from a simulated high pressure−high temperature (HPHT) reservoir was studied by Davani et al.[6] and Denney,[7] using numerical reservoir simulation methods, showing that a − 10 % error in gas viscosity can produce a relative 8.22 % error in estimated cumulative gas production, and a + 10 % error can lead to a relative 5.5 % error in cumulative production. Therefore, accuracy on viscosity data should have a large economical impact. The wide range of possible natural gas mixtures, depending basically on the origin, age, and depth of the reservoir,[8] and of conditions of interest, especially considering the HPHT conditions found in many new reservoirs that can be explored with current technologies,[9] precludes obtaining the relevant data by experimental studies on an exclusive basis. Moreover, to obtain accurate measurements of gas viscosity, especially for HPHT conditions, is very difficult, from a experimental viewpoint, and expensive.[6,10] Therefore, the analysis of published viscosity data found in the open literature shows that it is very limited in terms of both experimental conditions and quantity (see Atilhan et al.[11] and references therein), and in some cases their accuracy is unknown. Thus, the common practice in the natural gas industry is to estimate viscosity from available correlations and predictive methods that are based on available laboratory data.[12−14] Nevertheless, the predictive methods commonly applied in the industry have a reduced range of applicability, both in pressure−temperature conditions and mixtures' composition, and thus their application for HPHT conditions or for mixtures not previously tested may be doubtful.[7,11,15] Therefore, the available viscosity models do not satisfy the current industrial requirements for flow assurance and reliable reservoir characterizations. The development of accurate and reliable natural gas viscosity models, or testing of

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Table 1. Composition of Mixtures Studied in This Work

	mole fraction x^a		
component	QNG-S3[b]	QNG-S4[c]	QNG-S5[d]
methane	0.803 40	0.847 00	0.850 94
ethane	0.051 89	0.055 84	0.055 29
propane	0.018 78	0.019 62	0.020 09
i-butane	0.003 84	0.004 16	0.004 01
n-butane	0.005 73	0.005 53	0.006 12
i-pentane	0.001 88	0.002 14	0.001 71
n-pentane	0.001 40	0.001 55	0.001 41
n-octane	0.001 45	0.001 50	0.001 52
toluene	0.000 92	0.000 98	
methylcyclopentane	0.000 92		0.000 99
nitrogen	0.065 96	0.037 11	0.034 96
carbon dioxide	0.043 80	0.024 57	0.022 96

^{a} Relative uncertainty: methane 0.2 %, C2 to C4 2.0 %, C5 plus higher 5 %, N_2 and CO_2 2 %. ^{b} x (C_{6+}) = 0.00329. ^{c} x (C_{6+}) = 0.00248. ^{d} x (C_{6+}) = 0.00251.

[handwritten annotation: 350K = 76.85 °C = 170.35 °F; 37.5 MPa = 5437.5 psi]

the available ones, stands on the availability of experimental data, measured for a wide pressure–temperature ranges, for mixtures selected in a systematic way to infer the effects of the different components on the mixtures' viscosity, which compositions represent relevant reservoirs. We report in this work experimental viscosity data obtained from an electromagnetic piston viscometer at pressures from (10 to 65) MPa and temperatures between (250 and 450) K, for three selected natural gas-like mixtures whose compositions are characteristic of North Field reservoir after sweetening. This work is a continuation of a previous one[11] and is part of an international multilaboratory research project on the study of the main thermophysical properties of Qatari-type natural gas mixtures. The objectives of the work are to obtain accurate viscosity data for Qatari-type gases that may be used for production, processing, and transportation purposes; to infer the accuracy/reliability of common predictive viscosity models by use of the obtained viscosity data as reference; and to analyze the effects of mixtures' composition on the gas viscosity.

Experimental Section

Materials. Synthetic natural gas samples compositions resemble those of typical sweetened Qatar North Field gas. The compositions of the three studied samples (QNG-S3 to QNG-S5) are reported in Table 1. Linde Inc. prepared them gravimetrically in Spain and certified them according to ISO 6142. Sample composition was confirmed in our laboratory before measurements by gas chromatography, leading to the uncertainties reported in Table 1.

Samples were selected to infer the effects of methylbenzene to methylcyclopentane and nitrogen to carbon dioxide on the mixtures' viscosity. The C_2 to C_5 fractions and the octane contents are almost the same for the three studied mixtures. Sample 1 (QNG-S3) contains almost double the quantity of CO_2 and N_2 and a lower quantity of methane in comparison with sample 2 (QNG-S4) and sample 3 (QNG-S5), and thus, from the behavior of this mixture we may infer the effect of CO_2 and N_2 on natural gas viscosity. QNG-S4 contains toluene but no methylcyclopentane and vice versa for QNG-S5, and thus, from these samples, the effect of aromatics and cyclic compounds on mixtures' viscosity may be inferred.

Measurement and Calibration Methodology. The experimental viscosity data reported in this work were obtained from an electromagnetic viscometer developed by Cambridge Viscosity Inc. and commercialized by Vinci Technologies (electromagnetic viscometer EV1000). The experimental methodology,

together with a description of the calibration methods, apparatus performance, and analysis of measurement uncertainty, were reported in a previous work,[11] and thus they will be described only briefly in this work. The viscometer is a linear device; therefore, only two different standard gas samples are necessary for calibration corresponding to fluids in the high and low ends of the measurement range. Propane gas sample was obtained from Linde Inc. as ultra high purity with a mole fraction of 0.9995. Nitrogen gas sample is obtained from Linde Inc. as ultra high purity with a mole fraction of 0.999 999. Viscosity data for propane has a relative uncertainty of ± 2.5 % (below 30 MPa) and relative uncertainty of ± 4 % (at pressures above 30 MPa) for the entire temperature range. Moreover, relative uncertainty of the viscosity data for nitrogen from NIST Chemistry Webbook is ± 2 % over the entire temperature and pressure ranges. Methane viscosity is measured within the ± 2 % uncertainty range, which is greater than the ± 1 % uncertainty value reported by the manufacturer; however, in our opinion, it is not possible to achieve this level of accuracy for gas viscosity measurements, in the pressure–temperature ranges studied, by use of this apparatus. The isothermal viscosity data reported in this work covers the temperatures from (250 to 450) K temperature range (in 10 K steps) and (10 to 65) MPa range (in 5 MPa steps), and thus, 252 experimental viscosity data points are reported for each sample.

Measurements are based in the movement of an electromagnetically driven stainless steel piston inside a sensor chamber containing the fluid under study. The piston moves at constant force between two magnetic coils with the traveling time being a function of the fluid's viscosity. The relationship between piston traveling time and sample viscosity is established through a calibration procedure, described in detail in a previous work,[11] in which, when it is considered that the viscometer is a linear device, only two different standards are necessary for calibration corresponding to fluids in the high and low ends of the measurement range. Therefore, propane with a mole fraction of 0.9995 and ultrapure nitrogen with a mole fraction of 0.999 999 were used for calibration with their reference data extracted from the literature.[16–18] The working equations relating traveling time to viscosity, as a function of pressure and temperature, were developed and described by Atilhan et al.[11] A built-in platinum resistance thermometer (uncertainty ± 0.01 K) and a pressure transducer (uncertainty ± 0.01 MPa full scale) provide temperature and pressure measurements, respectively. The temperature and pressure sensors were calibrated through traceable procedures.[11] Therefore, the reproducibility of the measurements is better than ± 0.1 %; nevertheless, calculated uncertainties are ± 2.5 % (below 30 MPa) and ± 4.0 % (above 30 MPa) because of the uncertainties rising from the viscosity data used for calibration fluids.

The studied samples contain remarkable quantities of heavy components, and thus they could condense in the measurement assembly during experiments. The characteristic points along the phase envelope (cricondentherm, cricondenbar, and critical point) for QNG-S3 to QNG-S5 samples were calculated from the Peng–Robinson equation of state (Table 2). Values reported in Table 2 should be considered as initial estimates; experimental measurements of these phase envelopes are being carried out in our laboratories and they will be reported in future works. Nevertheless, calculated cricondentherm temperatures for the three studied samples are above common ambient conditions, and thus, to avoid condensation, sample cylinders were heated and kept above 350 K for at least 2 weeks before measurements.

Table 2. Calculated Cricondentherm, Cricondenbar, and Critical Point of Studied Samples[a]

	cricondentherm		cricondenbar		critical point	
	T/K	P/MPa	T/K	P/MPa	T/K	P/MPa
QNG-S3	303.43	4.09	257.34	11.09	213.31	7.59
QNG-S4	302.58	3.94	255.53	10.69	211.49	6.92
QNG-S5	302.58	3.91	255.83	10.66	211.80	6.92

[a] All values were obtained from the Peng−Robinson equation of state.

Moreover, all tubing lines and pumps used for gas compression were maintained above 350 K during the experiments.

Results

Measurements. Experimental viscosity data are reported in Table 3 (for sample QNG-S3), Table 4 (for sample QNG-S4), and Table 5 (for sample QNG-S5). Viscosity increases with increasing pressure along isotherms for the three studied mixtures and for all the temperatures, as expected. Nevertheless, the rate of change of viscosity with pressure is remarkably dependent both on the mixture characteristics and on the temperature, as shown in Figure 1. The three studied samples show almost the same viscosity, at fixed temperature and pressure, for pressures lower than around 30 MPa. This is an important conclusion for gas transportation purposes, when it is considered that the typical operating pressures of gas pipelines are lower than 20 MPa, and hence, flow of the three studied samples through pipelines should be almost the same for isobaric/isothermal conditions. Nevertheless, the behavior of QNG-S4 and QNG-S5 samples is almost parallel but completely different than the behavior of QNG-S3 sample at more than 30 MPa. QNG-S3 sample contains almost double the quantity of N_2 and CO_2, and a lower quantity of methane, than QNG-S4 and QNG-S5. Therefore, increasing concentrations of N_2 and CO_2 lead to a viscosity increase, which is in agreement with previously reported results.[11] The effect of non-hydrocarbon gases increasing the viscosity of the gas mixture is known in the literature, and in fact corrections including this effect were applied for classical viscosity correlation methods such as the Carr−Kobayashi−Burrows method;[19] nevertheless, the molecular origins of this behavior are still not fully clarified. Analysis of available literature viscosity data for methane + N_2[20] and methane + CO_2[21,22] binary mixtures shows that, for both systems, the evolution of viscosity with mixture composition leads to higher values (positive deviations) than the additivities of viscosities of corresponding pure compounds. The molecular origin of this viscosity behavior may be inferred from the available experimental/theoretical studies for the methane + N_2 system, based on related transport properties such as diffusion coefficients that have showed how the interaction between both molecules is weak;[23] this fact has been confirmed by computational studies of the interaction potential between methane and N_2 molecules.[24] In the case of methane + CO_2 system, weak interactions between both molecules could be inferred from available results,[25] which would justify the role of CO_2 molecules increasing mixture viscosity upon addition to methane-rich gases. Moreover, methane + N_2 or CO_2 binary systems show type I phase behavior[26−28] according to the van Konynenburg classification, which is a sign of weakly interacting molecules.

The effect of toluene and methylcyclopentane on mixtures' viscosity seems to be almost the same: QNG-S4 contains toluene and no methylcyclopentane, and vice versa for QNG-S5; however, the viscosity values, and their variation with pressure and temperature, for both samples are almost the same.

Table 3. Experimental Dynamic Viscosity, η,[a] of QNG-S3 Synthetic Natural Gas-like Mixture as a Function of Pressure, P, and Temperature, T

	T/K																				
P/MPa	250.00	260.00	270.00	280.00	290.00	300.00	310.00	320.00	330.00	340.00	350.00	360.00	370.00	380.00	390.00	400.00	410.00	420.00	430.00	440.00	450.00
10.0					14.67	14.57	14.55	14.59	14.67	14.79	14.92	15.08	15.24	15.42	15.61	15.80	16.00	16.21	16.41	16.62	16.83
15.0	26.00	23.43	21.53	20.16	19.18	18.49	18.01	17.67	17.45	17.31	17.24	17.21	17.23	17.27	17.34	17.43	17.54	17.66	17.79	17.93	18.08
20.0	32.45	29.61	27.29	25.42	23.94	22.79	21.88	21.19	20.65	20.24	19.94	19.71	19.55	19.44	19.37	19.34	19.34	19.37	19.41	19.48	19.55
25.0	37.52	34.56	32.06	29.96	28.22	26.79	25.61	24.64	23.86	23.23	22.72	22.31	21.98	21.73	21.53	21.38	21.27	21.20	21.15	21.14	21.14
30.0	41.85	38.78	36.15	33.90	31.99	30.38	29.01	27.86	26.90	26.09	25.42	24.85	24.39	24.00	23.69	23.43	23.22	23.06	22.93	22.83	22.76
35.0	45.76	42.57	39.81	37.44	35.39	33.64	32.13	30.84	29.73	28.79	27.98	27.29	26.71	26.21	25.79	25.43	25.14	24.89	24.69	24.52	24.38
40.0	49.41	46.09	43.21	40.70	38.53	36.65	35.02	33.61	32.39	31.33	30.41	29.61	28.93	28.33	27.82	27.38	27.00	26.68	26.40	26.17	25.98
45.0	52.89	49.43	46.41	43.78	41.49	39.49	37.75	36.23	34.90	33.74	32.72	31.83	31.05	30.37	29.78	29.26	28.81	28.42	28.08	27.78	27.53
50.0	56.27	52.66	49.50	46.74	44.32	42.21	40.35	38.73	37.30	36.04	34.93	33.95	33.09	32.33	31.67	31.08	30.56	30.10	29.70	29.35	29.04
55.0	59.59	55.81	52.50	49.61	47.06	44.83	42.87	41.14	39.61	38.26	37.06	36.01	35.07	34.23	33.50	32.84	32.26	31.74	31.28	30.88	30.52
60.0	62.88	58.93	55.46	52.42	49.74	47.39	45.31	43.48	41.86	40.42	39.14	38.00	36.98	36.08	35.27	34.56	33.91	33.34	32.83	32.37	31.97
65.0	66.17	62.02	58.39	55.19	52.38	49.91	47.72	45.78	44.06	42.53	41.16	39.95	38.86	37.89	37.01	36.23	35.53	34.90	34.34	33.84	33.38

[a] Values are given in units of μPa·s.

Table 4. Experimental Dynamic Viscosity, η,[a] of QNG-S4 Synthetic Natural Gas-like Mixture as a Function of Pressure, P, and Temperature, T

												T/K									
P/MPa	250.00	260.00	270.00	280.00	290.00	300.00	310.00	320.00	330.00	340.00	350.00	360.00	370.00	380.00	390.00	400.00	410.00	420.00	430.00	440.00	450.00
10.0					14.52	14.40	14.19	14.31	14.47	14.65	14.85	15.05	15.27	15.49	15.72	15.95	16.18	16.42	16.65	16.89	17.13
15.0	26.10	23.49	21.53	20.11	19.09	18.37	17.33	17.13	17.02	16.99	17.01	17.07	17.16	17.28	17.41	17.56	17.72	17.90	18.08	18.26	18.45
20.0	32.34	29.54	27.23	25.36	23.86	22.69	21.13	20.55	20.13	19.83	19.63	19.50	19.43	19.40	19.42	19.46	19.53	19.62	19.73	19.84	19.98
25.0	37.20	34.32	31.87	29.80	28.08	26.65	24.99	24.12	23.42	22.88	22.46	22.15	21.91	21.74	21.62	21.55	21.51	21.51	21.53	21.58	21.64
30.0	41.35	38.38	35.82	33.63	31.76	30.17	28.64	27.56	26.67	25.93	25.33	24.85	24.46	24.15	23.91	23.72	23.59	23.49	23.43	23.39	23.39
35.0	45.09	42.01	39.35	37.05	35.06	33.35	32.02	30.79	29.75	28.87	28.13	27.51	27.00	26.57	26.21	25.92	25.69	25.51	25.36	25.26	25.18
40.0	48.58	45.39	42.61	40.19	38.10	36.27	35.15	33.81	32.66	31.67	30.82	30.09	29.47	28.93	28.48	28.10	27.79	27.52	27.30	27.13	26.99
45.0	51.91	48.59	45.69	43.16	40.95	39.02	38.07	36.64	35.40	34.32	33.38	32.56	31.85	31.23	30.70	30.24	29.85	29.51	29.23	28.99	28.78
50.0	55.14	51.68	48.65	46.00	43.67	41.64	40.82	39.32	38.00	36.84	35.82	34.92	34.14	33.45	32.85	32.32	31.86	31.46	31.12	30.82	30.56
55.0	58.32	54.70	51.53	48.75	46.31	44.16	43.42	41.85	40.46	39.23	38.15	37.19	36.34	35.59	34.92	34.34	33.82	33.36	32.96	32.61	32.31
60.0	61.47	57.69	54.36	51.45	48.88	46.62	45.91	44.27	42.82	41.53	40.38	39.36	38.46	37.65	36.93	36.29	35.72	35.22	34.77	34.37	34.02
65.0	64.62	60.65	57.17	54.11	51.42	49.04	48.28	46.59	45.08	43.73	42.53	41.46	40.50	39.64	38.87	38.19	37.57	37.02	36.53	36.09	35.70

[a] Values are given in units of μPa·s.

Table 5. Experimental Dynamic Viscosity, η,[a] of QNG-S5 Synthetic Natural Gas-like Mixture as a Function of Pressure, P, and Temperature, T

												T/K									
P/MPa	250.00	260.00	270.00	280.00	290.00	300.00	310.00	320.00	330.00	340.00	350.00	360.00	370.00	380.00	390.00	400.00	410.00	420.00	430.00	440.00	450.00
10.0							14.33	14.31	14.47	14.65	14.85	15.05	15.27	15.49	15.72	15.95	16.18	16.42	16.66	16.89	17.13
15.0	26.05	23.45	21.50	20.08	19.07	17.97	17.30	17.10	17.00	16.97	17.00	17.06	17.15	17.27	17.40	17.56	17.72	17.89	18.07	18.26	18.45
20.0	32.27	29.48	27.18	25.32	23.83	22.16	21.07	20.50	20.08	19.79	19.59	19.47	19.40	19.38	19.40	19.44	19.51	19.60	19.71	19.83	19.96
25.0	37.11	34.24	31.80	29.75	28.04	26.44	24.90	24.04	23.36	22.82	22.41	22.10	21.86	21.70	21.58	21.51	21.48	21.48	21.50	21.55	21.61
30.0	41.23	38.28	35.74	33.56	31.70	29.87	28.52	27.45	26.57	25.85	25.26	24.78	24.40	24.09	23.86	23.67	23.54	23.45	23.39	23.36	23.35
35.0	44.95	41.90	39.25	36.96	34.99	33.29	31.86	30.65	29.63	28.76	28.03	27.42	26.91	26.49	26.14	25.86	25.63	25.45	25.31	25.21	25.13
40.0	48.42	45.25	42.49	40.10	38.01	36.20	34.95	33.64	32.50	31.53	30.69	29.97	29.36	28.83	28.39	28.02	27.71	27.45	27.23	27.06	26.92
45.0	51.73	48.44	45.56	43.04	40.85	39.35	37.84	36.44	35.21	34.15	33.22	32.42	31.72	31.11	30.59	30.14	29.75	29.42	29.14	28.90	28.71
50.0	54.95	51.51	48.50	45.87	43.56	41.98	40.56	39.08	37.78	36.64	35.64	34.76	33.98	33.31	32.71	32.20	31.74	31.35	31.01	30.72	30.47
55.0	58.10	54.51	51.36	48.60	46.18	44.69	43.13	41.58	40.22	39.01	37.94	37.00	36.16	35.42	34.77	34.19	33.68	33.24	32.84	32.50	32.20
60.0	61.23	57.48	54.18	51.29	48.74	47.17	45.58	43.97	42.55	41.28	40.15	39.15	38.26	37.46	36.76	36.13	35.57	35.07	34.63	34.24	33.90
65.0	64.36	60.42	56.97	53.93	51.26	49.71	47.93	46.26	44.78	43.46	42.28	41.22	40.28	39.43	38.68	38.00	37.40	36.86	36.37	35.94	35.56

[a] Values are given in units of μPa·s.
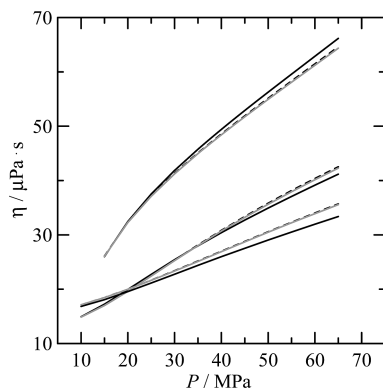
Figure 1. Comparison of isothermal viscosity data for QNG-S3 to QNG-S5 samples. From top to bottom: data for 250 K, 350 K, and 450 K. QNG-S3, solid black lines; QNG-S4, dashed black lines; QNG-S5, solid gray lines. Guiding lines are plotted, experimental points are omitted for the sake of clarity.


Figure 2. Temperature at which viscosity minimum appears along isobars. QNG-S3, solid black line; QNG-S4, dashed black line; QNG-S5, solid gray line. Lines show linear fits to experimental values.

Moreover, if we compare viscosity data previously reported for the mixture QNG-S1,[11] whose composition is almost the same as QNG-S4 and QNG-S5 but contains both toluene and methylcyclopentane, we may infer that viscosity for these three samples is almost the same. Hence, the effect of considering mixtures with only toluene, with only methylcyclopentane, or with both compounds seems to be the same for viscosity for a fixed concentration. Therefore, we may conclude that aromatics and cyclic compounds should have the same effect on a mixture's viscosity as shown in Figure 1. Literature viscosity data for the methane + toluene binary system[29] show lower values than the additivity of values for pure compounds. Moreover, the analysis of viscosity data for methane + toluene system points to a certain degree of associative interaction between both molecules,[29] and thus this fact, together with the remarkable asymmetric character of the mixture, would justify the lower viscosities for the binary system and the effect of toluene on the viscosity of the multicomponent mixtures studied in this work. To our knowledge, viscosity data for the methane + methylcyclopentane system are not available in the open literature; nevertheless, the behavior of this system should be very similar to that of methane + toluene one, considering that both binary mixtures show type III behavior according to the van Konynenburg classification, and thus, a certain degree of association between toluene or methylcyclopentane and methane molecules could be inferred.[30]

The temperature effect upon viscosity along isobars is dependent on pressure. For lower pressures, viscosity evolution with temperature shows minima, whereas for larger pressures, viscosity decreases with increasing temperature; this behavior was obtained for the three studied samples. The temperature for which the viscosity minima appear shifts to higher values as the pressure increases as shown in Figure 2. These trends may be explained by considering that for low pressures and high temperatures a low-density gas is obtained (studies of density for these mixtures are being performed and will be published in future works), and thus, the regular trend for gases of increasing viscosity with increasing temperature is obtained. For high pressures, a compressed gas is obtained, being remarkably denser than for low-pressure conditions, and thus liquidlike viscosity behavior, viscosity decreasing with increasing temperature, is obtained. The behavior for intermediate pressures is also intermediate, and thus leads to the minima reported in Figure 2.[31] Results reported in Figure 2 show that the behavior of QNG-S4 and QNG-S5 is almost the same and is completely different than QNG-S3; the minima for QNG-S3
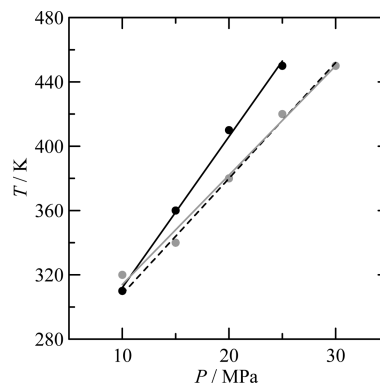
disappear for lower pressures in comparison with QNG-S4 and QNG-S5. This is in agreement with the higher density of QNG-S3 sample in comparison with QNG-S4 and QNG-S5 samples, leading to a gaslike viscosity pressure−temperature region extending over shorter pressure−temperature ranges for QNG-S3 in comparison with QNG-S4 and QNG-S5 systems, as inferred from Figure 2 results.

Analysis of Predictive Ability for Selected Viscosity Models. Experimental measurement of viscosity for complex gas mixtures, in wide pressure−temperature ranges, is a complex task, and thus, when the very different compositions of natural gases are considered, it is neither possible nor practical to do measurements for all the possible systems. Therefore, development and testing of theoretical models for viscosity prediction purposes is required by the natural gas industry. Several models have been developed and analyzed in the literature against highly accurate data,[15,32−35] and even new models have been proposed recently for mixtures with characteristics not included in the previously available models (higher contents of heavier alkanes, inorganic gases, sour gases).[36,37] In this work, we have selected four viscosity models commonly used in the natural gas industry, because of their simplicity and supposed reliability, and compared their predictions with the experimental viscosity data reported in this work for the three studied mixtures. The models selected are NBS (Nilpotent−Bazhanov−Stroganov) developed by Ely and Hanley[38] at NIST, also known as TRAPP (latest version is SUPERTRAPP); CLS (Chung−Lee−Starling);[39,40] LBC (Lohrenz−Bray−Clark);[41] and PFCT (Pedersen−Fredenslund−Christensen−Thomassen).[42,43] The main characteristics of the four models were briefly described in a previous work.[11]

A comparison between experimental and predicted results at 250 K, 350 K, and 450 K and (10 to 65) MPa appears in Figure 3 and in Table 6, in which the average error of each model for each sample is reported. The deviations between experimental and predicted results of the models follow the trend NBS > CLS > LBC > PFCT. Moreover, deviations are larger for the QNG-S3 sample (richer in N_2 and CO_2) than for QNG-S4 and QNG-S5 (containing lower quantities of N_2 and CO_2) for all the studied models, pressures, and temperatures. Therefore, as we reported in a previous work,[11] the effect of increasing N_2 and CO_2 concentration on mixture viscosity is not properly described by the studied models, leading to larger deviations. It should be remarked that model deviations obtained in this work for QNG-S4 and QNG-S5 samples are very similar to those obtained for the QNG-S1 sample reported in a previous work;[11] viscosity for QNG-S1 sample is almost the same as for QNG-S4 and QNG-S5 samples, and thus, the models' performance is very
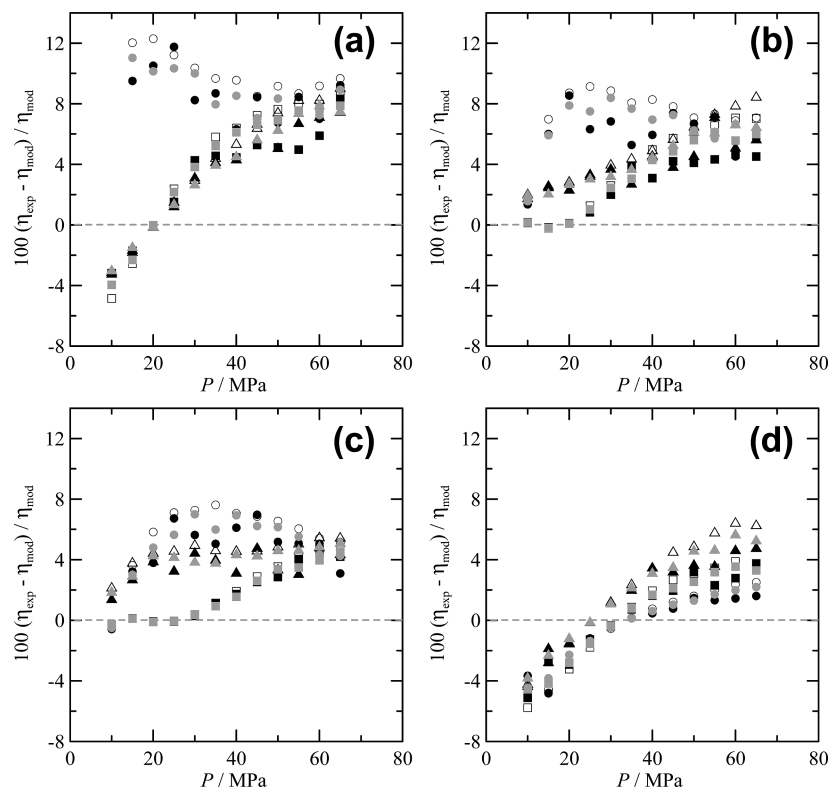
Figure 3. Percentage deviations between experimental, η_{exp}, and predicted, η_{mod}, viscosity values. Symbols: circles, values for 250 K; squares, values for 350 K; and triangles, values for 450 K. Open symbols, values for QNG-S3; black solid symbols, values for QNG-S4; and gray solid symbols, values for QNG-S5. (a) NBS, (b) CLS, (c) LBC, and (d) PFCT.

Table 6. Percentage Absolute Average Deviations between Experimental and Predicted Viscosity Data Obtained by Use of the Reported Models

	NBS	CLS	LBC	PFCT
QNG-S3	6.70	5.40	4.13	2.89
QNG-S4	5.57	4.21	3.40	2.32
QNG-S5	5.96	4.72	3.65	2.50

similar for all these samples. Moreover, although deviations are larger for QNG-S3 sample than for QNG-S1, QNG-S4, and QNG-S5 samples, the differences between them are not as large as the differences from sample QNG-S2 reported in a previous work[11] QNG-S2 sample did not contain eeither N₂ or CO₂, and the performance of all the models was clearly better than for the remaining studied mixtures. Therefore, N2 and CO₂ have a remarkable effect on the models' predictive ability. Nevertheless, deviations obtained with the PFCT model are remarkably lower than those obtained with the remaining models for all the studied mixtures, leading to viscosity predictions reasonably accurate over the whole pressure−temperature ranges analyzed. TRAPP, which is the older version of currently used SUPERTRAPP,[44] which was developed by NIST and can be used to calculate the thermodynamic and transport properties of pure fluids and fluid mixtures, might also yield better predictions as it has superior prediction capability than TRAPP.

Conclusion

This paper reports new experimental viscosity data for three synthetic natural gas-like mixtures, resembling gases from the Qatari North Field, covering the (250 to 450) K and (10 to 65) MPa temperature and pressure ranges, from an electromagnetic viscometer. The effect of N₂ and CO₂ content on the mixture viscosity was analyzed, showing that increasing concentration of these compounds tends to increase viscosity. Moreover, the

effects of aromatics and cyclic hydrocarbons were also analyzed by use of toluene and methylcyclopentane as models; results show analogous effects of both compounds leading to a decrease in viscosity. Predictive ability of four common viscosity models were analyzed in comparison with reported experimental viscosity data. Mixtures rich in N₂ and CO₂ lead to larger deviations for all the studied models; nevertheless, PFTC model shows a clearly superior performance, leading to deviations within the uncertainty levels of the experimental data reported in this work.

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